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Revision 0

CORROSION MONITORING IN HANFORD NUCLEAR WASTE STORAGE TANKS, DESIGN AND DATA FROM 241-AN-102 MULTI-PROBE CORROSION MONITORING SYSTEM

Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management

Contractor for the U.S. Department of Energy
Office of River Protection under Contract DE-AC27-08RV14800

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ABSTRACT

In 2008, a new Multi-Probe Corrosion Monitoring System (MPCMS) was installed in double-shell tank 241-AN-102 on the U.S. Department of Energy's Hanford Site in Washington State. Developmental design work included laboratory testing in simulated tank 241-AN-102 waste to evaluate metal performance for installation on the MPCMS as secondary metal reference electrodes. The MPCMS design includes coupon arrays as well as a wired probe which facilitates measurement of tank potential as well as corrosion rate using electrical resistance (ER) sensors. This paper presents the MPCMS design, field data obtained following installation of the MPCMS in tank 241-AN-102, and a comparison between laboratory potential data obtained using simulated waste and tank potential data obtained following field installation.

Keywords: tank potential, electrical resistance, corrosion monitoring, nuclear waste tank corrosion

INTRODUCTION

The U.S. Department of Energy's Hanford Site is located in the southeastern portion of Washington State. The Hanford Site has a total of 177 waste tanks, 28 "newer" double-shell waste tanks and 149 older single-shell tanks as well as many miscellaneous storage facilities. Together, the single-shell and double-shell waste tanks currently store approximately 57 million gallons of nuclear waste from the Hanford Sites weapons production mission. Retrieval projects are ongoing to remove waste from the older single-shell tanks and transfer it to the "newer" double shell tanks to be stored until the Vitrification Plant is online and able to process the stored nuclear waste ultimately, into glass.

The Hanford Site double-shell tanks (DSTs) were constructed of welded mild steels, including American Society for Testing and Materials (ASTM) A537 CL1 [Unified Number System (UNS) K02400], ASTM A515 Grade 60 (UNS K02401), and ASTM A516 Grade 60 (UNS K02100). The DST inner tanks were stress relieved at the completion of construction. The DSTs currently store supernatant

liquid and two possible different types of solid materials, sludge or saltcake, depending on the processes and original waste sources. The Hanford Site currently employs a chemistry-based corrosion mitigation program for the DSTs, which mainly uses Sodium Hydroxide (hydroxide) to adjust the pH of tank waste. The amount of hydroxide added is also dependent on the concentration of nitrates. Over time, the chemistries of several of the Hanford Site DSTs have shifted outside allowable chemistry specifications. Efforts to remediate non-compliant tanks are underway, but have not yet been completely successful.

In 2004, the Hanford Site began exploring modifications to the chemistry-based corrosion mitigation system through laboratory testing in waste simulants. The laboratory testing confirmed the importance of the waste tank corrosion potential (Ecorr) in regards to the onset of stress corrosion cracking (SCC). The Hanford Site is conducting an extensive laboratory testing program, under the oversight of a panel of industry and academic corrosion experts. This program is underway to develop recommendations for modified acceptable waste chemistry specifications, based on keeping the waste tanks from reaching corrosion potentials capable of inducing SCC.

The addition of hydroxide to the Hanford Site DSTs to maintain them within the corrosion mitigation program chemistry limits incurs costs associated with additional waste requiring long term disposal. Through the laboratory test program and assistance from the expert panel oversight committee, the Hanford Site has been successful in reducing the specification for DSTs that have been outside the corrosion mitigation program chemistry limits for extended periods of time. Tanks that have been recommended for reduced waste chemistry specifications are also monitored with "real-time" corrosion monitoring systems.

Several generations of corrosion monitoring systems have been installed in the Hanford Site DSTs; however, none of the previous instruments have been specifically designed and used to measure waste tank corrosion potential. In 2007, ARES Corporation was tasked to design a corrosion monitoring system for one of the Hanford Site DSTs, Tank 241-AN-102 (AN-102). Development of the AN-102 Multi-Probe Corrosion Monitoring System (MPCMS) required a multi-discipline design team as well as completion of laboratory testing in simulated tank waste. The design and fabrication support effort for the AN-102 MPCMS occurred during 2007 and 2008, culminating with MPCMS installation in AN-102 on the Hanford Site in May 2008. This paper presents the AN-102 MPCMS design, field data obtained following installation of the system, and a comparison between laboratory potential data obtained using simulated waste and tank potential data obtained following field installation.

SYSTEM DESIGN

In February 2007, an initial design specification for the AN-102 MPCMS was developed by the Hanford Site.[1] The MPCMS includes a fixed probe (designed to remain in the tank for monitoring throughout the operational life of the system) plus four removable probes (designed to be removed for examination at set intervals over the operational life of the system). The removable probes contain retrievable stressed and un-stressed corrosion coupons. The fixed probe contains standard and secondary reference electrodes as well as sets of stressed and unstressed coupons. All coupons are fabricated from the same material as tank AN-102 [i.e.,

ASTM A537 CL1 (UNS K02400A) steel]. A revision to the specification was submitted in May 2007 to include electrical resistance (ER) sensors at three locations on the fixed probe.[2] For simplicity, both specifications require data be collected by operators using a portable data collection device with hand-held leads.

The overall purpose of the AN-102 MPCMS is to enhance the Hanford Site's ability to measure and predict corrosion in the vapor space (the headspace above the tank liquid), the supernatant liquid, and the solids (saltcake) region of the tank. The stressed and unstressed coupons on the removable probes, although not monitored, will be weighed and examined upon removal to establish general corrosion rate and the propensity of the tank waste to induce pitting and SCC.

To facilitate installation in the tank, the four MPCMS removable probes are arranged in a circle surrounding the fixed probe. The entire five-probe assembly is installed as a single piece of equipment and must fit within an 11-in (28-cm) diameter circle. The riser in the tank dome for MPCMS installation is a 12-in (30.5-cm) riser. The probes are approximately 55-ft (16.8 m) in length and reach to within approximately 6-in (15.2-cm) of the tank bottom. The MPCMS design includes an integral spray ring assembly to facilitate rinsing of the removable probes and the fixed probe during removal.

Fixed Probe Design

The fixed probe includes the structures to support each of the four removable probes. The main portion of the fixed probe, the body, is a 2-in (5-cm) ASTM A106 (UNS K02501) Schedule 80 carbon steel pipe, abrasive blasted to National Association of Corrosion Engineers (NACE) #1/Society for Protective Coatings (SSPC)-SP5 white metal finish requirements, then primed and coated with epoxy (to facilitate decontamination upon removal) prior to electrode and coupon installation. The probe body is sealed on the bottom by a welded, rounded tip. The overall probe body length is approximately 55-ft (16.8 m). An overall view of the completed fixed probe assembly is presented in Figure 1. Arrays of instruments and coupons are included in the vapor space, the supernatant, and the saltcake. For simplicity, only ER sensors, reference electrodes, and weight loss coupons are used on the fixed probe. All electrode and ER sensor wiring terminates in a terminal box at the top of the MPCMS assembly (Figure 2).

The AN-102 MPCMS ER sensors are adapted from commercially-available sensors made out of American Iron and Steel Institute (AISI) 1020 Carbon Steel (UNS G1020). Ideally, these sensors would have been fabricated from ASTM A537 (UNS K02400) carbon steel to best approximate the corrosion behavior of the tank wall. However, from a general corrosion rate perspective, the two materials are expected to behave similarly. Each ER sensor on the 241-AN-102 MPCMS uses a measurement span of 0.005 in (0.0127-cm).

The unprocessed output of the ER sensors ranges from 0 to 1,000, with each unit equivalent to 0.1% of the measurement span in the ER sensor under consideration. Thus, each unit of raw data output is equivalent to the loss of 5E-6 in. (1.3E-5-cm) of wall thickness in the ER sensor. The AN-102 MPCMS design requires that ER sensor measurements are manually made at specified frequencies by field personnel. A view of an ER sensor installed on the fixed probe is presented in Figure 3.

To facilitate corrosion potential measurements, three types of primary reference electrodes are included in the fixed probe design. The primary reference electrodes are designed to withstand and operate at temperatures up to 160°F (71°C), pH ranges from 7 to 14, and radiation fields up to 200 (Rads) R/hr. Previous work by Danielson on five types of primary reference electrodes exposed to gamma radiation was consulted for preliminary reference electrode design.[3] Primary reference electrode types tested by Danielson included two types of silver/silver chloride reference electrodes, a calomel reference electrode, and two metallic reference electrodes, a platinum flag and a piece of ASTM A537 (UNS K02400) carbon steel. The testing indicated that the commercial-grade primary reference electrodes subjected to radiation failed after a short time. While the metallic secondary reference electrodes were able to better withstand the radiation, they experienced a shift in potential due to the radiation field.

Two sets of three primary reference electrodes were included in the AN-102 MPCMS design, one set in the supernatant, and one set in the saltcake. Traditional silver/silver chloride and calomel primary reference electrodes, plus a prototype hybrid double-junction copper sulfate/silver chloride primary reference electrode* were designed and built by Van London pHoenix Co. These specially-built electrodes are constructed out of polyvinylidene fluoride (PVDF) with porous junctions made of PVDF for radiation resistance. Figure 3 presents a view of the three electrode types.

The primary reference electrode bodies were not subjected to radiation testing during AN-102 MPCMS design development. Because these electrodes will eventually fail due to degradation by radiation or by electrolyte contamination, secondary reference electrodes were included in the MPCMS design. The ultimate goal of the inclusion of primary and secondary reference electrodes is to allow continued tank corrosion potential measurements following the eventual failure of the primary reference electrodes.

To select secondary reference electrode materials to be used on the MPCMS, laboratory testing was performed to characterize the stability of a variety of candidate secondary reference electrode materials. Testing was performed at Oak Ridge National Laboratory (ORNL) in 2007. The laboratory tests were performed to evaluate the stability of corrosion potentials over time and to characterize the open-circuit corrosion potentials of the candidate materials when exposed to high pH waste simulants. No radiation fields were imposed upon the specimens. The test setup utilized two different AN-102 liquid tank waste simulants, one at pH 11 and one at pH 14, at approximately 80°F (27 °C). Metals evaluated included silver rod, chloridized silver wire, platinum wire, nickel rod, copper rod, titanium rod, and Hastelloy C22®**, (UNS N06022) rod.

The secondary reference electrode open circuit corrosion potentials were measured against three different types of commercially-available primary reference electrodes: silver/silver chloride, calomel, and copper/copper sulfate reference electrodes. The laboratory test results indicated that most of the candidate materials exhibited relatively stable open circuit corrosion potentials. Based on performance and cost, copper, nickel, and silver were selected for use as

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* The hybrid double-junction copper sulfate/silver chloride primary reference electrode is essentially constructed as a double-junction silver/silver chloride reference electrode, but uses a 1 molar copper sulfate solution (instead of saturated potassium chloride) between the primary and secondary junctions.

** Hastelloy C22® is a registered trademark of Haynes International in Kokomo, Indiana.

secondary reference electrodes on the MPCMS.[4] One of each type of metallic secondary reference electrodes was installed in the supernatant and saltcake areas of the MPCMS. The secondary reference electrodes are electrically isolated from the carbon steel probe body by a glass seal. The secondary reference electrodes, as installed on the fixed probe, are shown in Figure 4.

Tank metal electrodes were also included in the design to facilitate corrosion potential measurements on the tank material of construction, ASTM A537 (UNS K02400). To best approximate the corrosion behavior of the tank wall, an archived plate of ASTM A537 (UNS K02400) of a similar vintage as the tank was used to fabricate the MPCMS tank metal reference electrodes and coupons. Prior to electrode/coupon fabrication, this plate was subjected to the same stress-relief anneal heat treatment applied to AN-102 following fabrication. The MPCMS design includes two bare tank metal reference electrodes installed at the same locations as the primary and secondary reference electrodes, i.e., one set in the supernatant and one set in the saltcake. One set of tank metal reference electrodes, as installed on the fixed probe, is shown in Figure 4.

Three types of coupons are used on the MPCMS: stressed, pre-cracked C-rings, round coupons, and bar-shaped coupons. The fixed probe contains three sets of stressed pre-cracked C-ring and round coupons; one set in the vapor space, one set in the supernatant, and one set in the saltcake. The primary function of the pre-cracked, stressed C-rings is to facilitate examination for the detection of SCC. The C-rings can also be used to provide weight loss data. The primary function of the round coupons is to provide weight loss data to establish uniform corrosion rate. The primary function of the bar-shaped coupons is to provide information on corrosion at the liquid/vapor interface. The C-ring coupons are fabricated and then pre-cracked until a crack is observed on both ends of the machined notch with the crack depth on one end between 0.020-in (0.051-cm) and 0.040-in (0.102-cm).[5] The C-rings are bolt-loaded to induce stress. A photograph of a bolt-loaded C-ring coupon is Figure 5. The bar-shaped coupon is 22-in (56-cm) long and will ultimately be positioned (after MPCMS installation) at the liquid/air interface (LAI) in the tank. The bar coupon will be examined to following removal to provide information on corrosion at the LAI. The coupons on the fixed probe are not monitored and will only be evaluated when the fixed probe body is removed from the tank at the end of the MPCMS service life. An installed set of C-ring and round coupons is presented in Figure 6.

The MPCMS fixed probe body additionally has an internal leak detection cable installed which is routed from the bottom of the probe to the terminal box installed at the top of the probe. Following leak detection cable installation, electrode and coupon installation, and leak testing, the probe body was filled with 2-lb density sprayable two-part closed-cell polyurethane foam. A piece of the leak detection cable was installed in a practice pipe and then foamed with the same two-part closed-cell polyurethane foam to determine leak detection cable capabilities. The AN-102 MPCMS factory acceptance testing indicated the cable should be capable of detecting a leak in the probe body even when embedded in the polyurethane foam.[6] The leak detection cable is new to the AN-102 MPCMS design and was integrated in the probe assembly based on experiences with the previous fiberglass reinforced plastic corrosion probe body used for the corrosion probe installed in DST 241-AN-107 in September 2006.[7]

Passive Probe Design

Four removable probes surround the fixed probe assembly. The removable probes are coupon racks that do not contain actively-monitored components. The removable probe bodies are fabricated out of ASTM A36 (UNS K02600) carbon steel 2 ½-in (6.35-cm) x 1 1/2-in (3.8-cm) x ¼-in (0.64-cm) angle iron, abrasive blasted to NACE #3/SSPC-SP6 commercial blast finish requirements, then primed and coated with epoxy prior to coupon installation. Each piece of angle iron is approximately 55-ft (16.8-m) in length. The same types of weight loss coupons installed on the fixed probe are installed on the removable probes. Three round and three pre-cracked stressed C-ring coupons are included in each tank area; the vapor space, the supernatant, and the saltcake, on each removable probe. One bar-shaped coupon is included on each of the removable probes. Coupons on the removable probes are mounted at elevations similar to those on the fixed probe. The removable probes will be removed individually from the tank on a specified frequency for coupon forensic inspection. The coupon elevations are stamped into the angle iron as a method of correlating the initial coupon weight to the final weight after tank exposure to allow corrosion rate calculation.

Additional MPCMS Design Features

Since the MPCMS is installed in a radioactive waste tank and equipment decontamination is a concern, the system includes a novel spray canister design to allow for decontamination of the individual removable probes and the fixed probe assembly during probe removal from the tank. Flaps and individual removable probe spray nozzles were integrated in the design to allow each probe to be rinsed prior to removal from the tank. Additionally, the holders for each of the angle iron removable probes were extended above the top flange to allow connection of contamination control sleeving during removal. These design features should prevent the top of the MPCMS system from becoming contaminated, a desirable goal as the equipment will be installed in the 241-AN Tank Farm for several years. For fixed probe assembly removal, the spray canister will remain mounted to the riser, and the whole probe body can be decontaminated as the probe is being removed from the riser.

Tank AN-102 contains a relatively-thin, but hard layer of saltcake (i.e., solid) waste that the probe assembly had to break through during installation. To facilitate installation, a water lance was incorporated into the MPCMS design to cut through the saltcake layer. The water lance pipe has a conical spray nozzle at the bottom capable of providing water at a rate of 27 gpm at 100 psi per the manufacturer. Additional water lance features included double check valves installed in the tank vapor space region to prevent the backflow of contaminated water through the water lance piping.

DATA

The AN-102 MPCMS was installed on May 1, 2008. Corrosion potential data from the supernatant region are presented in Figures 7 through 9. Figures 10 through 12 present corrosion potential data from the tank saltcake region. For clarity, spurious data from human errors in the data collection process have been removed from the figures. A plot of metal loss data and associated corrosion rate data from the ER sensors is shown in Figure 13.

DISCUSSION

Figures 7 and 10 show corrosion potential data for the tank and tank material electrodes relative to the primary reference electrodes in the supernatant and saltcake regions of the tank, respectively. Previously-performed laboratory work has shown that SCC is unlikely at corrosion potentials more negative than approximately -100 mV (vs. Saturated Calomel Electrode).[8] Based on a comparison of the data in Figures 7 and 10 with laboratory test data, the corrosion potentials of the tank and tank material electrodes are well below (more negative) than the most negative corrosion potential shown to induce SCC in the laboratory work.

With the exception of the hybrid copper sulfate/silver chloride primary reference electrode in the saltcake [denoted as "$CuSO_4$ (AgCl)" in the figures], data indicate that all electrodes are relatively stable and functioning as expected. The sudden shifts in the corrosion potentials of the tank and saltcake tank material electrodes relative to the saltcake $CuSO_4$ (AgCl) primary reference electrode starting on May 13, 2008, are not likely indicative of a sudden change in waste corrosivity since no concurrent shifts were recorded in the potential of other electrodes in the same region (i.e., a large change in corrosion conditions within the tank should impact numerous electrodes). The shift appears to be indicative of a stand-alone problem or malfunction of the saltcake $CuSO_4$ (AgCl) primary reference electrode (e.g., junction failure in the electrode, contamination of the electrical connection to the electrode with the graphite-based thread sealant, and/or contamination of the electrode filling solution). The mode of failure can only be assumed for now since the in-tank MPCMS cannot be easily removed. The malfunction of the saltcake $CuSO_4$ (AgCl) electrode is not expected to impact the integrity of the in-tank MPCMS, the ability to collect corrosion potential data from the other electrodes, or the ability to measure uniform corrosion rate via the ER sensors.

Figures 8 and 11 show corrosion potential data for the tank and tank material electrodes relative to the secondary reference electrodes in the supernatant and saltcake regions of the tank, respectively. Figures 9 and 12 show voltage differences between the primary and secondary reference electrodes in the supernatant and saltcake regions of the tank. Though primary reference electrode data from the tank are presently available and being used to monitor waste corrosivity, secondary reference electrode data are being collected and monitored to both verify that the secondary reference electrode potentials remain relatively stable over time, and to facilitate the transition from primary to secondary reference electrode data following the eventual failure of the primary reference electrodes.

In addition to corrosion potential electrodes, the fixed probe also contains three ER sensors; one in the vapor space, one in the supernatant, and one in the saltcake layer of the tank. Data from the ER sensors are shown in Figure 13. All ER sensors indicate uniform corrosion rates of well under 1 mpy (0.025 mmpy), the design corrosion allowance for the tank. Raw metal loss data from the ER sensors is used to calculate uniform corrosion rate (also shown in Figure 13). To smooth the output, the corrosion rate data shown in Figure 13 have been processed to show a rolling 10-day average corrosion rate. By averaging data over a 10-day period, data plots are smoothed while maintaining good responsiveness to changes in corrosion conditions.

Overall corrosion rates have also been calculated for each of the ER sensors by performing a regression analysis on the metal loss data over the time period evaluated. The results are then converted to a corrosion rate in mils per year (mpy). The overall corrosion rate is not as responsive to immediate changes in tank waste corrosivity, but is useful for estimating overall wall thickness loss from year-to-year. The overall corrosion rates from each of the three ER sensors are presented in Table 1.

TABLE 1: ER SENSOR OVERALL CORROSION RATES.

ER Sensor	**Overall Corrosion Rate (mpy) / (mmpy)**
Vapor Space	-0.0048 / -1.2E-4
Supernatant	0.0002 / 5E-6
Saltcake	-0.0012 / -3E-5

As is evident in Table 1, corrosion rates being recorded are at the limit of detection of the instrumentation and essentially fluctuate around zero (leading to the occasional "negative" overall corrosion rate).

The instrument used to read the ER sensors, has a published metal loss repeatability of ± 1 division, where 1 division is equal to 0.005 mil and "metal loss repeatability" is the capability of the ER instrument to measure the same value regardless of the length of time between the measurements, assuming the value to be measured has not changed. Error associated with the metal loss repeatability in divisions can be converted to error in corrosion rate based on the frequency at which ER sensor measurements are made. The likelihood of obtaining the same measured value if the value to be read has not changed increases as a function of the time period between measurements. For example, the ER sensors on the 241-AN-102 MPCMS are currently measured every 7 days; therefore, the error associated with the metal loss repeatability is ± 1 division over a 7 day period, or ±0.26 mpy (0.0066 mmpy). Conversion of the metal loss repeatability rate from divisions/day to mpy is performed by multiplying the division/day rate by 0.005 mil/division and 365 day/year. Thus, for example, in the event the ER sensor measurement frequency was increased to once per day, the error associated with the metal loss repeatability would be ± 1 division over 1 day, or ± 1.83 mpy (0.046 mmpy).

It is important to note that reporting the metal loss repeatability rate on a yearly basis is not reasonable if only two data points are obtained at a close interval; for example, if the ER sensor data were taken every 2 minutes, the associated metal loss repeatability error would indicate an error rate of ± 1314 mpy (33.4 mmpy). The fact that regression analysis of a multiplicity of data points taken over an extended period of time does not produce a significant positive slope is an indication that general corrosion rate is zero (0 mpy) or very close to zero (0 mpy).

Readings from the ER sensors to date are within the error range for metal loss repeatability (± 1 division or ± 1.8 mpy (0.046 mmpy)), and at the limit of detection of the instrumentation (i.e., corrosion rates are essentially 0 mpy). The "negative" corrosion rates evident in Table 1 are an artifact of the fact that ER readings are fluctuating around 0 mpy, not

an indication of plating or other electrochemical reactions capable of increasing the thickness of the ER sensors over time.

SUMMARY AND CONCLUSIONS

The MPCMS consists of a fixed probe surrounded by four removable probes in an assembly that fits within a 12-in (30.5-cm) riser and was specifically designed for installation in Tank 241-AN-102. The MPCMS was installed and the first readings to monitor the waste tank potential were taken on May 1, 2008.

With the exception of the $CuSO_4$ (AgCl) primary reference electrode in the saltcake layer of the tank, which appears to be malfunctioning, data to date indicate that all other electrodes on the fixed probe are functioning properly. Based on comparison with laboratory test data, the corrosion potentials recorded on the tank material electrodes and tank wall are well below (more negative than) the most negative corrosion potential shown to induce SCC in the laboratory work. The failure of the saltcake $CuSO_4$ (AgCl) primary reference electrode is not expected to impact the integrity of the MPCMS or other data collection capabilities of the system.

All ER sensors on the MPCMS appear to be functioning as designed. Readings from the ER sensors to date are within the error range for metal loss repeatability [± 1 division or ± 1.8 mpy (0.046 mmpy)], and at the limit of detection of the instrumentation (i.e., corrosion rates are essentially 0 mpy).

No removable probes have been removed from the tank to date. Thus, no coupon weight loss or forensic examination data are available for presentation.

The AN-102 MPCMS facilitates the monitoring of tank corrosion potential and corrosion rate, metrics critical to the effective management of this tank, particularly as the tank approaches or exceeds its design life. The system is expected to provide an unprecedented level of information on tank waste corrosivity, and should become a valuable asset for effective DST management at the Hanford Site.

ACKNOWLEDGMENTS

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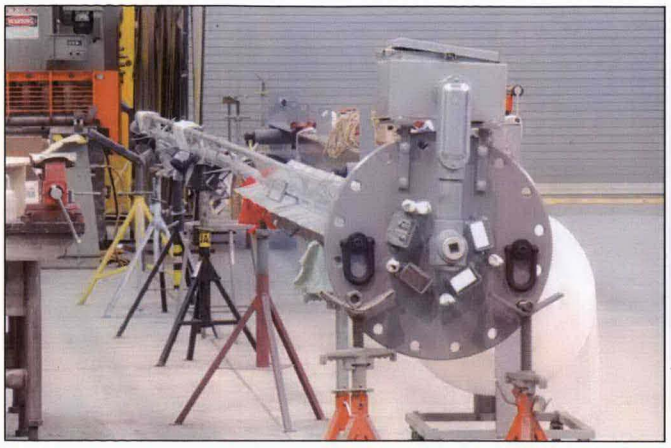

FIGURE 1 – Active Probe Overview

FIGURE 2 – Terminal Box at Top of MPCMS

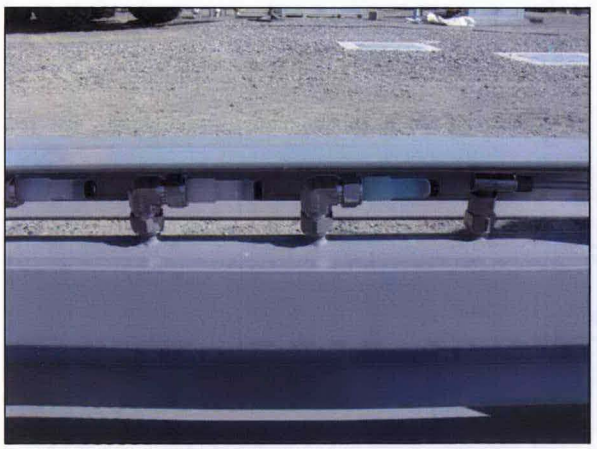

FIGURE 3 – Installed Primary Reference Electrodes and ER Sensor

FIGURE 4 – Secondary Reference Electrodes

FIGURE 5 – Bolt-Loaded C-Ring Coupon

FIGURE 6 – ASTM A537 C-Ring and Round Coupons

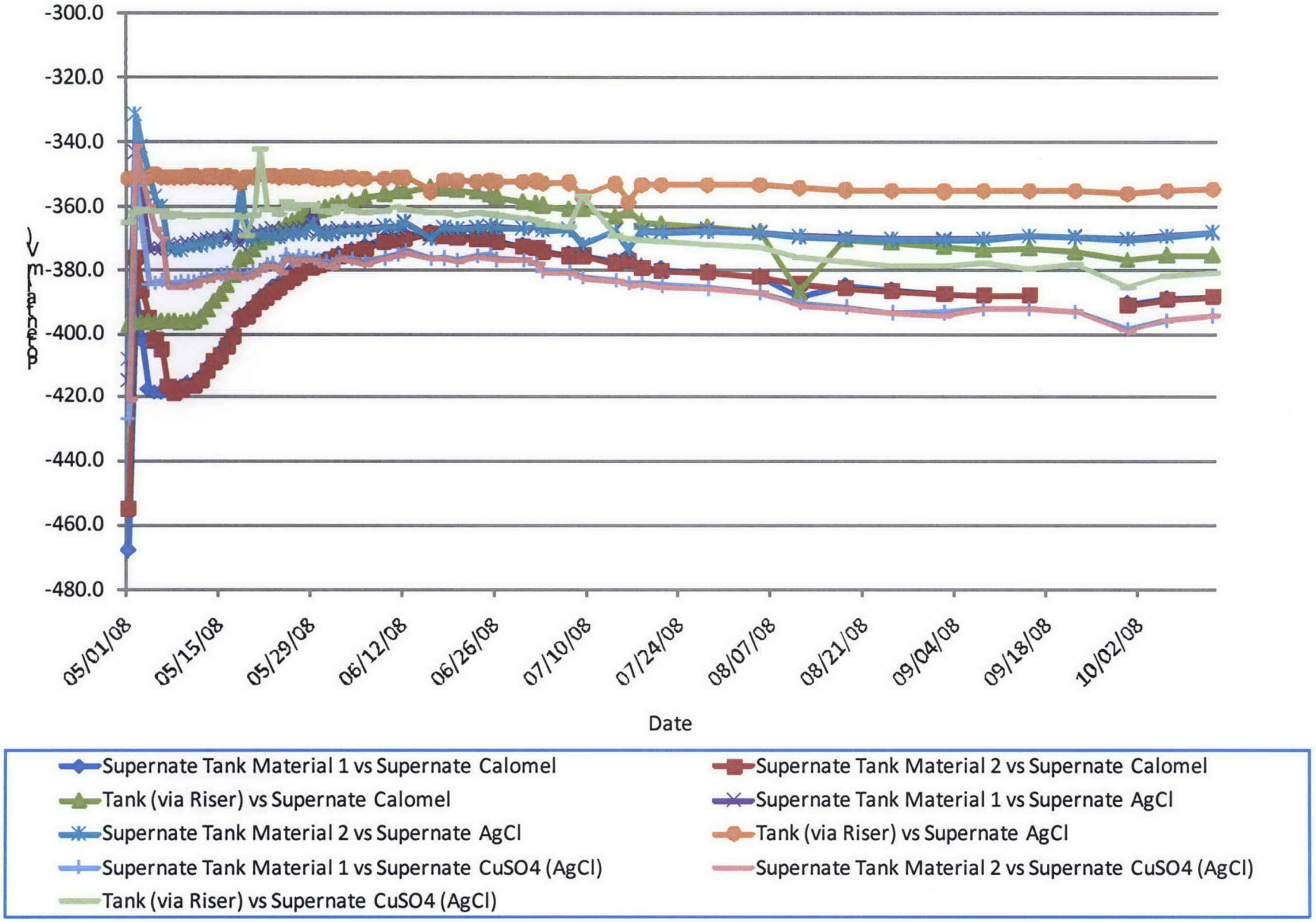

FIGURE 7 - Supernatant Tank Material vs. Primary Reference Electrodes

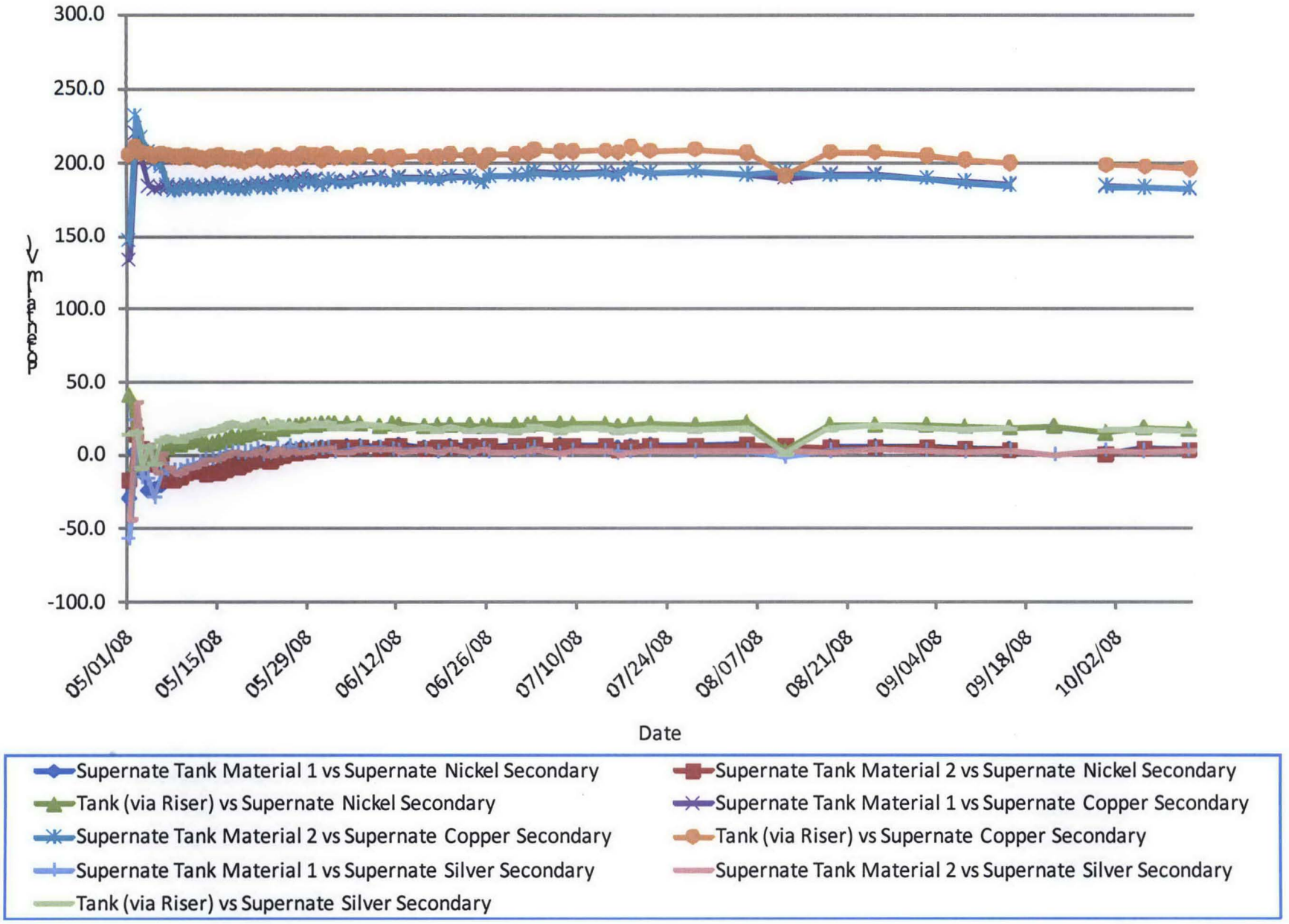

FIGURE 8 - Supernatant Tank Material vs. Secondary Reference Electrodes

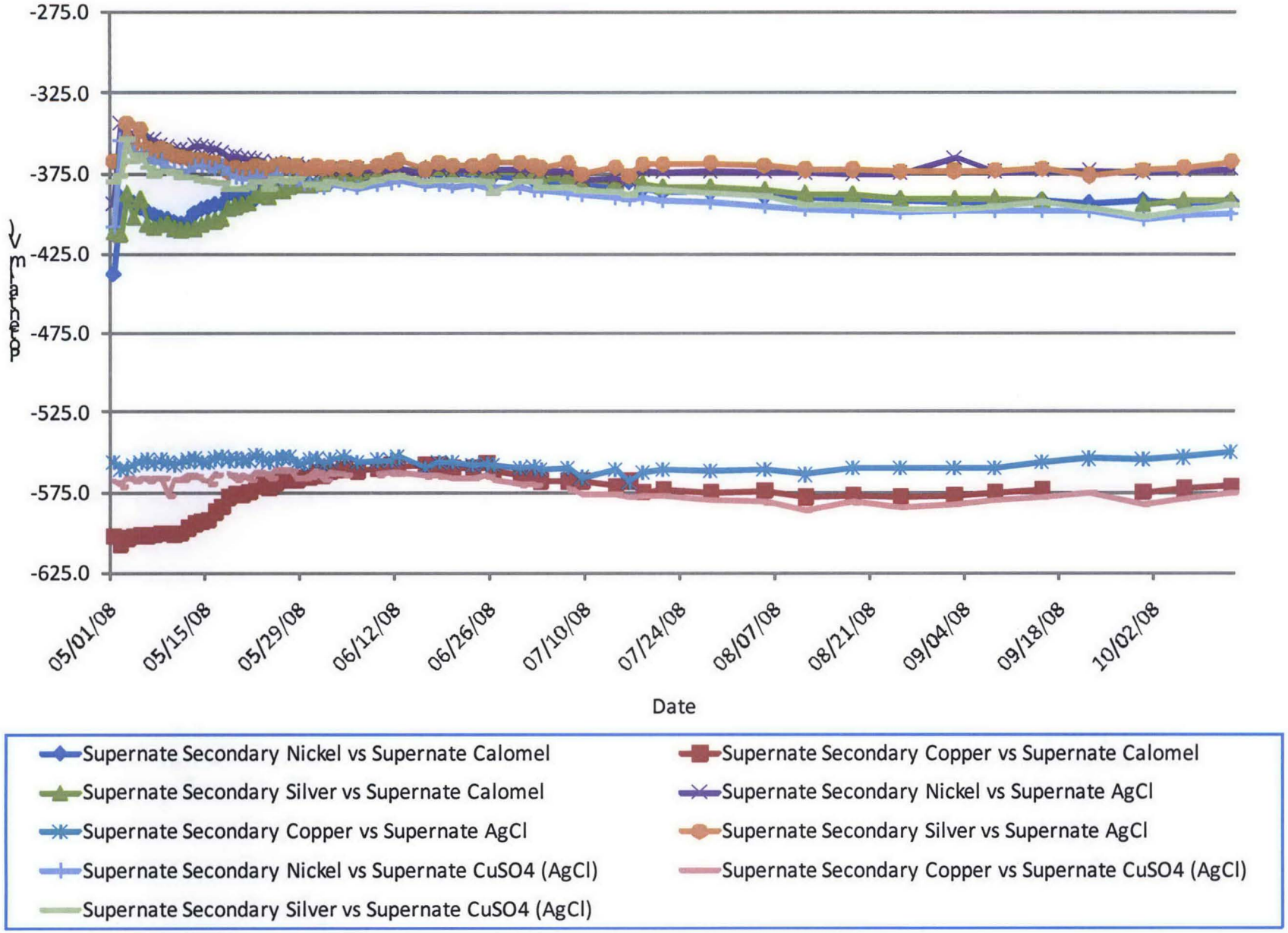

FIGURE 9 - Supernatant Secondary Reference vs. Primary Reference Electrodes

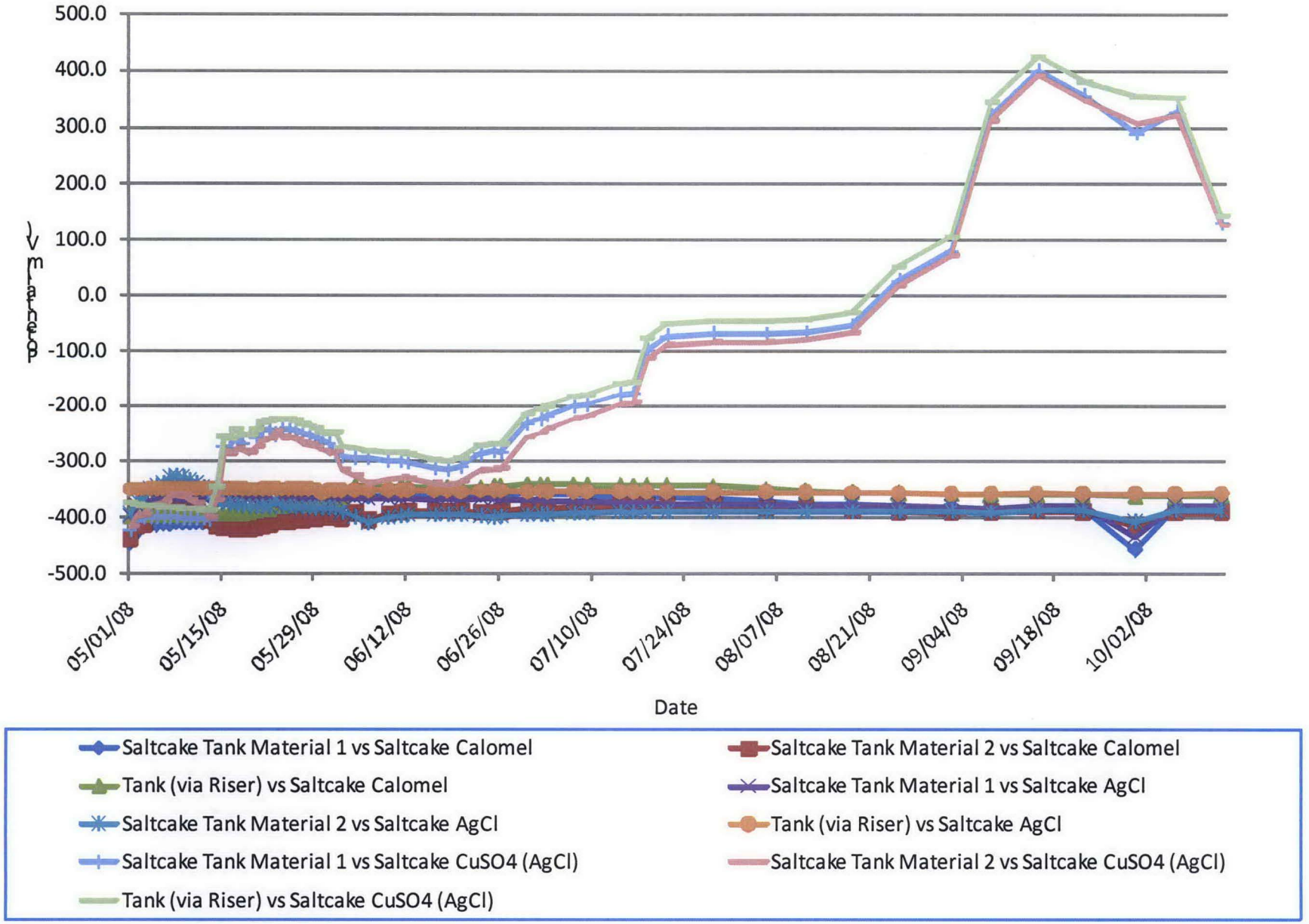

FIGURE 10 – Saltcake Tank Material vs. Primary Reference Electrodes

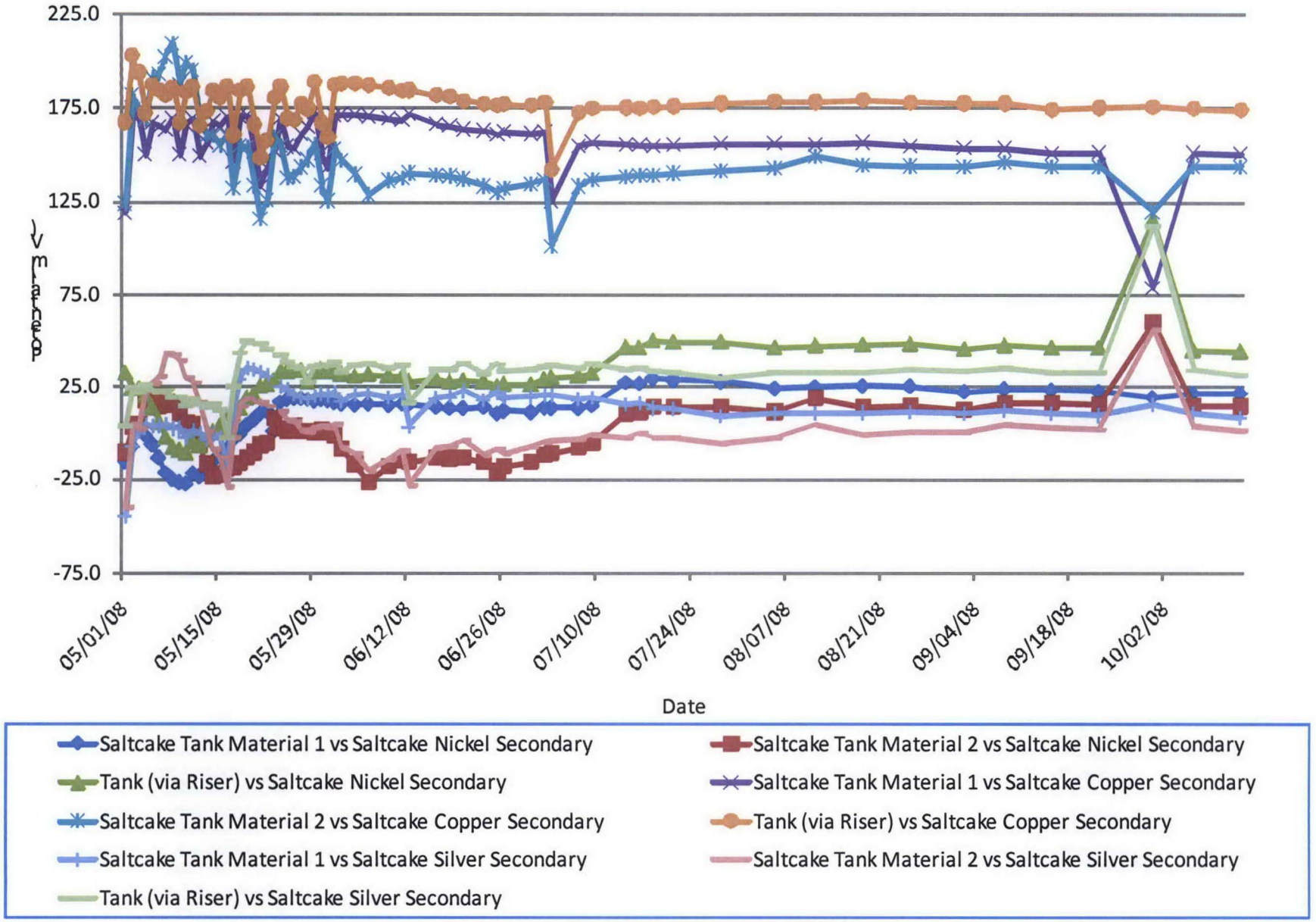

FIGURE 11 - Saltcake Tank Material vs. Secondary Reference Electrodes

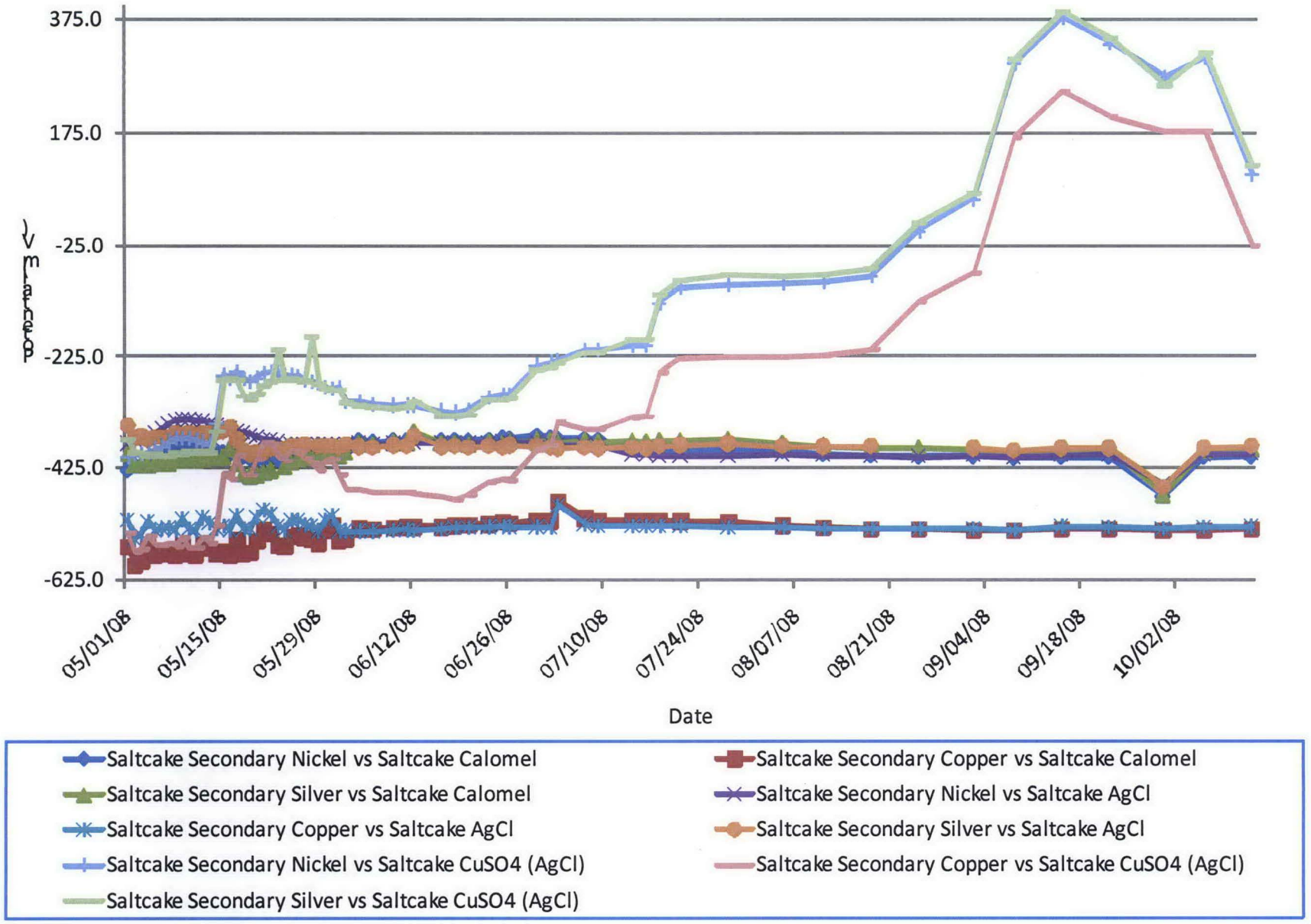

FIGURE 12 – Saltcake Secondary Reference Electrodes vs. Primary Reference Electrodes

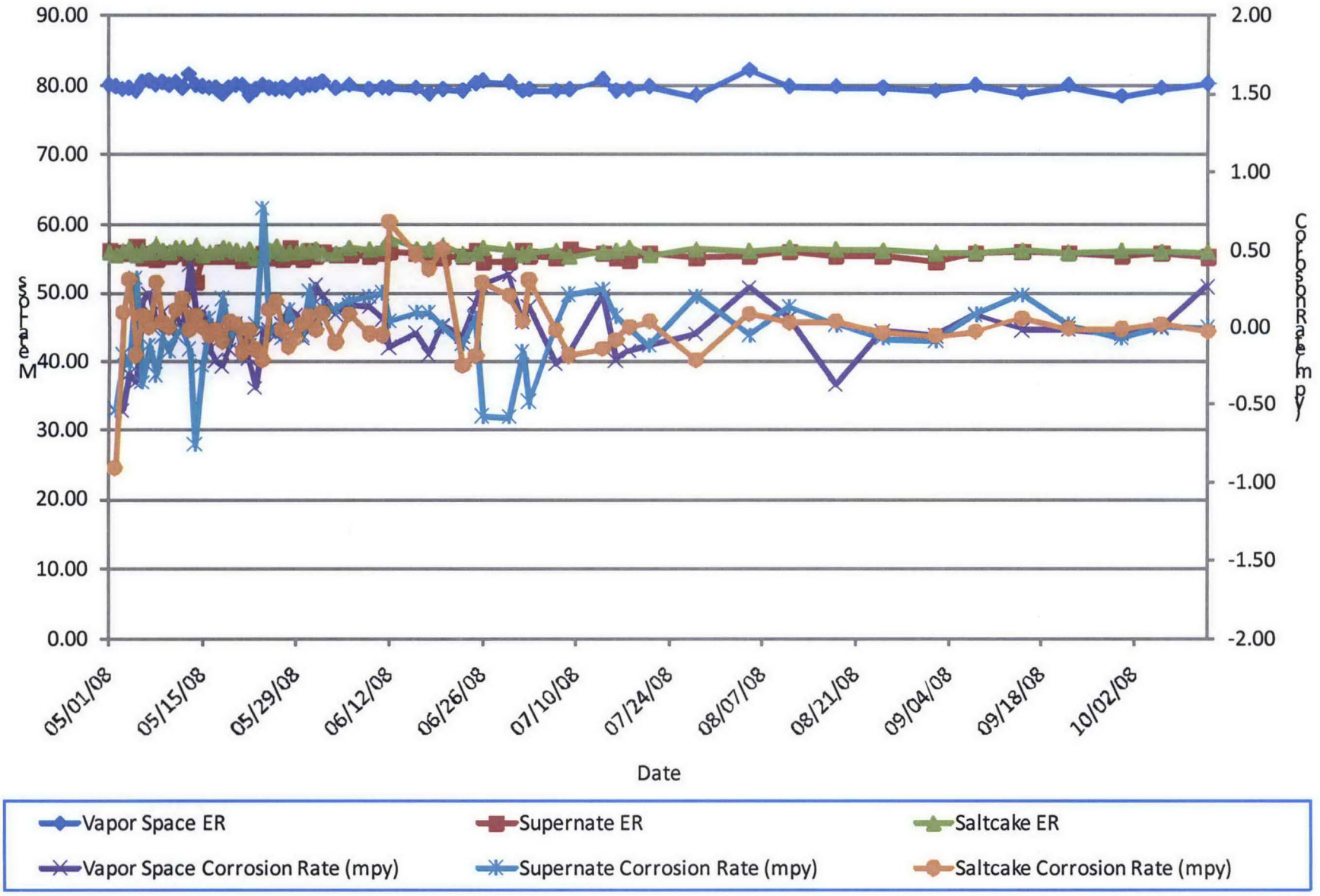

FIGURE 13 – ER Sensor Metal Loss and Calculated Corrosion Rates